

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference PCT66-DC4511	<b>FOR FURTHER ACTION</b> See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/CA98/00875	International filing date (day/month/year) 17/09/1998	Priority date (day/month/year) 07/11/1997
International Patent Classification (IPC) or national classification and IPC C08F255/02		
Applicant FISH, Robert, Benham, Jr. et al.		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.



2. This REPORT consists of a total of 7 sheets, including this cover sheet.

☒ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of 5 sheets.

3. This report contains indications relating to the following items:

- I ☒ Basis of the report
- II ☐ Priority
- III ☒ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- IV ☐ Lack of unity of invention
- V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI ☐ Certain documents cited
- VII ☒ Certain defects in the international application
- VIII ☒ Certain observations on the international application

Date of submission of the demand  27/05/1999	Date of completion of this report  14.02.99
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International application No. PCT/CA98/00875

**I. Basis of the report**

1. This report has been drawn on the basis of (*substitute sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to the report since they do not contain amendments.*):

**Description, pages:**

1-19,21-25	as originally filed			
20	as received on	25/01/2000	with letter of	20/01/2000

**Claims, No.:**

1-17	as received on	25/01/2000	with letter of	20/01/2000
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2. The amendments have resulted in the cancellation of:

- ☐ the description, pages:
- ☐ the claims, Nos.:
- ☐ the drawings, sheets:

3. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

4. Additional observations, if necessary:

**III. Non-establishment of opinion with regard to novelty, inventive step and industrial applicability**

The questions whether the claimed invention appears to be novel, to involve an inventive step (to be non-obvious), or to be industrially applicable have not been examined in respect of:

- ☐ the entire international application.
- ☒ claims Nos. 2.

because:

- ☐ the said international application, or the said claims Nos. relate to the following subject matter which does not require an international preliminary examination (*specify*):

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- ☐ the description, claims or drawings (*indicate particular elements below*) or said claims Nos. are so unclear that no meaningful opinion could be formed (*specify*):
- ☒ the claims, or said claims Nos. 2 are so inadequately supported by the description that no meaningful opinion could be formed.
- ☐ no international search report has been established for the said claims Nos. .

**V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

**1. Statement**

Novelty (N)	Yes:	Claims	1, 3-17
	No:	Claims	
Inventive step (IS)	Yes:	Claims	
	No:	Claims	1,3-17
Industrial applicability (IA)	Yes:	Claims	1, 3-17
	No:	Claims	

**2. Citations and explanations**

**see separate sheet**

**VII. Certain defects in the international application**

The following defects in the form or contents of the international application have been noted:

**see separate sheet**

**VIII. Certain observations on the international application**

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

**see separate sheet**

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Reference is made to the following documents:

- D1: EP-A-0 027 375 (TOA NENRYO KOGYO KK) 22 April 1981  
D2: WO 95 16750 A (DU PONT ;HAYASHI RYUICHI (JP); KOSHIDA REIKO (JP)) 22 June 1995  
D3: US-A-5 451 639 (MARCZINKE BERND L ET SEQ. AL) 19 September 1995  
D4: 'GRAFTED HETEROPHASIC POLYMERS FOR THE IMPACT MODIFICATION OF NYLON' RESEARCH DISCLOSURE, no. 358, 1 February 1994, page 84 XP000439817

**Re item III**

For the figure 0.9 in the claim 2, received 25 January 2000, a basis in the original documents was neither specified, nor could it be found (Art. 19(2), 34(2)b PCT).

**Re item V**

**Novelty**

In D1 unusual high Melt flow indices (MI) values are disclosed (page 10, lines 1-5) apparently due to the use as laminating resin.

In the application the upper limit of MI is 5 g/min (claim 1). Thus, the subject matter of the claims is novel over D1.

D2 - D4 relate to the same technical field as the application namely toughener compositions for polyamides on the basis of blends of two olefine copolymers that are cografted with olefinic carboxylic acids or their anhydrides, typically maleic acid anhydride

However:

In D2 the densities are not mentioned in D2. Moreover (claim 1) component (a) is a cyclic poly olefin, whereas in the application component (a) is a copolymer of ethylene and an  $\alpha$ -olefin having at least 4 carbon atoms.

In D3 a mixture of two propylene copolymers is cografted.

According to D4 a blend of of an ethylene- propylene rubber (b) in a propylene-ethylene copolymer or a propylene C<sub>4-10</sub>  $\alpha$ -olefin copolymer (a) is cografted.

According to the application (a) is a copolymer of ethylene and an  $\alpha$ - olefin having at

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least 4 carbon atoms. Also the densities are not mentioned.

Thus, novelty (Art. 33(2) PCT) is given.

**Inventive step**

Since D1 refers to a another technical field, it cannot be used as starting point for the assessment of inventive step.

However, it is unclear which technical problem is solved by the respective distinguishing features (e.g. selection of densities) in a non obvious way. Thus, it is unclear as to whether and how inventive step might be established over D2 - D4.

On the basis of the evidence submitted, it appears doubtful as to whether or not to problem to avoid massing is linked with the respective distinguishing features. The authors of D2 to D4 do not report that they encountered massing problems nor had they to employ a partitioning agent (as formulated in the application on page 2, l. 23). This is in line with the explanation in the description on page 4, lines 13-15: "In particular, the use of the ethylene  $\alpha$ -olefin in pre-grafted blend eliminates the need for either a pre-modification or post-modification partitioning agent." Since also in D2 to D4 the (rubbery) elastomers were used in combination with ethylene  $\alpha$ -olefin copolymers, it appears to be logical that also the authors of D2 to D4 did not encounter a general problem of massing.

In another formulation: it appears that the problem was already solved in D2 to D4 without this being stated explicitly.

**Independent process claim 14 and process claims 15 to 17**

The grafting of a blend of two olefin polymers (in overlapping amounts) in an extruder at an overlapping barrel temperature (50 -295°C, page 10, lines 13-17) and the blending of the cografed polyolefins with polyamide is disclosed in D2, e.g. claims 1 and 3; this refers to claims 13, 14, 16.

D2 further discloses polyamide 6 and 66 (p. 3, l. 19 and 22) and the absence of a partitioning agent; this refers to claims 1, 15 and 17.

Thus the process features as such are not novel or well known in the art.

**Novelty** of the process is established by component (a) of the starting materials (cf.. the analysis as to claim 1).

Since it appears that the use of the component (a) as claimed ( i.e. the distinguishing feature) does not involve **inventive step**, the same applies for the process as a whole.

All claims: Industrial applicability (Art. 33(4) PCT) is given.

### **Re item VII**

With the amended version of page 20S, received on 25 January 2000, in example 27 the value for dryflow was amended from 1.09 to 109. This amendment introduces subject-matter which extends beyond the content of the application as filed, contrary to Article 19(2) PCT.

### **Re Item VIII**

#### **Certain observations on the international application**

As to the feature "**massing**" the following is noted:

This is a unusual feature: cf.. PCT Preliminary Examination Guidelines, Chapter III, 4.7a " *The examiner should be aware of the possibility that applicants may attempt to employ unusual parameters to disguise lack of novelty.*"

The term "massing" is a relative term only and thus, obscure. In the description on page 20 it explained " the massing tendencies were evaluated by subjecting the polymer under a pressure of 100 g/ cm<sup>2</sup> for a period of 24 hrs. ... At the end of the period, the polymer was inspected to see if the pellets have massed together and if so, whether the mass can be broken up."

This is not a clear definition since no boundaries are given.

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The feature "massing" may be tolerable in the claims for purely illustrative purposes; the feature, however, cannot be used for defining the subject matter of the application (Art. 6 PCT, clarity; PCT Preliminary Examination Guidelines, Chapter III, 4.5).

As to "toughening agent useful for improving the impact properties of polymeric compositions" (claims 1, 9, 14):

"toughening agent " represents an attempt define the subject matter of the claims or a part thereof by the the result to be achieved. This normally is not admissible (Art. 6 PCT; PCT Preliminary Examination Guidelines, Chapter III, 4.7).

Since "useful for" must be construed as meaning merely " suitable for", such a passage normally does not represent a limitation (Art. 6 PCT; PCT Preliminary Examination Guidelines, Chapter III, 4.8).

The extremely broad content of the passage in the **description** on page 16, line 24 to page 17, line 8 (here just only a blend of "at least one polymer and PE") is in contradiction with the claims (Art. 6 PCT).

**Claim 11:** (any) "graft monomer" is not supported by the description ( Art 6 PCT). -

It appears that any chemical product is somehow "a fabricated material "; cf.. Collins English Dictionary, key word "fabricate" = "to make, build, devise, invent". Thus, this term in the context given is devoid of any further meaning.

The expression "such as" has no limiting effect, the feature following ("moulded automobile parts") should be regarded as entirely optional (Art. 6 PCT; PCT Preliminary Examination Guidelines, Chapter III, 4.6).

Table 1

	Co-graft Example	Composition			MI (190C) dg/min.	MA %	Dryflow sec	Massing 50C, 24 hr.
		EPR%	PE type	PE%				
5	1	78	PE 1	22	4.6	0.92	36	50C, 24 hr.
	2	76	PE 1	24	3	1.2	22	none
	3	47.5	PE 1	52.5	0.95	1	5.7	none
	4	47.5	PE 1	52.5	1.7	0.89	5.8	none
	5	19	PE 1	81	0.9	0.82	5.5	none
10	6	19	PE 1	81	0.49	1.27	5.8	none
	7*	0	PE 1	100	0.43	1.41	5.4	none
	8*	0	PE 1	100	0.43	0.92	5.3	none
	9	76	PE 2	24	4.1	0.96	18	none
	10	76	PE 2	24	3.2	1.38	16	none
15	11	47.5	PE 2	52.5	2	1.2	7.6	none
	12	47.5	PE 2	52.5	2.8	0.84	8.3	none
	13	19	PE 2	81	1.5	0.91	6.2	none
	14	19	PE 2	81	1.1	1.33	6.3	none
	15	0	PE 2	100	1	1.4	6.4	none
20	16	0	PE 2	100	1.21	0.91	6.4	none
	17*	100		0	6.9	0.67	48	massed
	18	76	PE 3	25	4.7	1.03	89	breakable
	19	76	PE 3	24	3.2	1.26	61	breakable
	20	47.5	PE 3	52.5	2.5	1.05	40	none
25	21	19	PE 3	81	3.8	0.89	31	none
	23	19	PE 3	81	2.3	1.17	28	none
	24*	0		100	2.4	1.24	30	none
	25*	0		100	3.6	0.91	34	none
	26	85.5	PE 3	14.5	5.6	1.08	82	massed, breakable
30	27	90	PE 3	10	9.1	0.84	109	massed
	28*	95		5	5.8	1.13	47	massed, breakable

\* = comparative

The massing tendencies were evaluated by subjecting the polymer under a pressure of 100 g/sq. cm at 50°C for a period of 24 hr. The pressure is similar to the one experienced by the bottom layer of a 1 ton pallet of resins. At the end of the period, the polymer was inspected to see if the pellets have massed together and if so, whether the mass can be broken up.

In the dry flow experiment, 250g of the resin was allowed to pass through a stemmed plastic funnel (diameter of the stem is about 2 cm). The time it took for the resin to pass through was recorded. This time is dependent on the shape of the pellets, but since all the samples are produced under similar conditions, their shapes are similar. The relative dry flow time is therefore a good indication of how easy the resin can be handled.



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WHAT IS CLAIMED IS:

1. A polymeric toughening agent useful for improving the impact properties of polymeric compositions, comprising,
  - 5 (a) a copolymer of ethylene with one or more  $\alpha$ -olefins having at least 4 carbon atoms and having a density of 0.930 to 0.880 g/cc and a melt index (MI) of 0.01 to 50 dg/min at 190°C, 2.16 Kg;
  - (b) a massing polymer selected from a copolymer of ethylene with one or more  $\alpha$ -olefins having at least 3 carbon atoms and having a density of  
10 0.850 to 0.880 g/cc and an MI of .01 to 50 dg/min at 190°C, 2.16 Kg wherein the ratio of (a) to (b) is 10:90-90:10; and
  - (c) .05-5 wt % relative to (a) and (b) of a grafted monomer covalently bonded to (a) and (b) selected from an olefinic carboxylic acid or anhydride or derivative thereof.
- 15 2. The polymeric toughening agent of claim 1, comprising 0.9-5 wt% of component (c).
3. The polymeric toughening agent of claim 1 wherein component (a) has a density of 0.890 to 0.920 g/cc and an MI of 0.1 to 10 dg/min and component (b) has a density of 0.855 to 0.875 g/cc and an MI of 0.1 to 10 dg/min.
- 20 4. The polymeric toughening agent of claim 1 wherein component (a) has a density of 0.890 to 0.920 g/cc and an MI of 0.1 to 10 dg/min and component (b) has a density of 0.855 to 0.875 g/cc and an MI of 0.1 to 10 dg/min and wherein component (c) is 0.1 to 3 wt % relative to (a) and (b).
5. The polymeric toughening agent of claim 1 wherein component (a) has a density  
25 of 0.90 to 0.910 g/cc and an MI of 0.5 to 5 dg/min and component (b) has a density of 0.86 to 0.87 g/cc and an MI of 0.2 to 2 dg/min.

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6. The polymeric toughening agent of claim 1 wherein component (a) has a density of 0.90 to 0.910 g/cc and an MI of 0.5 to 5 dg/min and component (b) has a density of 0.86 to 0.87 g/cc and an MI of 0.2 to 2 dg/min and wherein component (c) is 0.3 to 2 wt % relative to (a) and (b).

7. The polymeric toughening agent of claim 1 wherein component (a) is selected from a linear low density polyethylene and component (b) is selected from an ethylene/propylene/diene monomer, ethylene/propylene rubber; a metallocene polyethylene having a melt flow ratio  $I_{10}/I_2$  of less than 6.53 and an  $M_w/M_n$  ratio of greater than the melt flow less 4.63; a metallocene polyethylene having a melt flow ratio  $I_{10}/I_2$  of less than 6.13 and an  $M_w/M_n$  ratio of equal or less than the melt flow ratio less 4.63 and component (c) is selected from the group consisting of acrylic acid, methacrylic acid, fumuric acid, maleic acid, nadic acid, citaconic acid, itaconic acid and anhydrides, metal salts, esters, amides or imides of said salts.

8. The polymeric toughening agent of claim 7 wherein the toughening agent is non-massing.

9. A polymeric composition having improved impact properties, comprising:

(1) a polymeric toughening agent useful for improving the impact properties of the polymeric composition, comprising,

(a) a copolymer of ethylene with one or more  $\alpha$ -olefins having at least 4 carbon atoms and having a density of 0.930 to 0.880 g/cc and an MI of 0.01 to 50 dg/min at 190°C, 2.16 Kg;

(b) a massing polymer selected from a copolymer of ethylene with one or more  $\alpha$ -olefins having at least 3 carbon atoms and having a density of 0.850 to 0.880 g/cc and an MI of .01 to 50 dg/min at 190°C, 2.16 Kg wherein the ratio of (a) to (b) is 10:90-90:10; and

(c) .05-5 wt % relative to (a) and (b) of a grafted monomer covalently bonded to (a) and (b) selected from an olefinic carboxylic acid or anhydride or derivative thereof; and

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(2) an olefinic or non-olefinic material.

10. The polymeric composition of claim 9 wherein the polymeric toughening agent comprises 0.09-5 wt% of component (c).

11. The polymeric composition of claim 9 wherein the non-olefinic material is  
5 selected from a polyamide and wherein the weight percentage ratio of (1):(2) is 3-40:97-60.

12 The polymeric composition of claim 11 wherein the polyamide is selected from nylon 6 or nylon 6,6 and wherein the weight percentage ratio of (1):(2) is 15-25:85-75.

13. A fabricated material such as molded automobile parts, comprising:

10 (1) a polymeric toughening agent useful for improving the impact properties of olefinic or non-olefinic materials, comprising,

(a) a copolymer of ethylene with one or more  $\alpha$ -olefins having at least 4 carbon atoms and having a density of 0.930 to 0.880 g/cc and an MI of 0.01 to 50 dg/min at 190°C, 2.16 Kg;

15 (b) a massing polymer selected from a copolymer of ethylene with one or more  $\alpha$ -olefins having at least 3 carbon atoms and having a density of 0.850 to 0.880 g/cc and an MI of 0.01 to 50 dg/min at 190°C, 2.16 Kg wherein the ratio of (a) to (b) is 10:90-90:10; and

(c) .05-5 wt % relative to (a) and (b) of a graft monomer covalently  
20 bonded to (a) and (b); and

(2) an olefinic or non-olefinic material.

14. A process for producing a polymeric composition having improved impact properties, comprising,

(a) preparing a non-massing toughening agent useful for improving the impact  
25 properties of the polymeric composition as claimed in Claim 1; and

(b) combining the non-massing toughening agent produced in step (a) with

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- (1) a polyamide to form, under suitable reaction conditions, the polymeric composition, or
- (2) another non-olefinic material or olefinic material to form the polymeric composition.

5 15. The process according to claim 14 wherein a partitioning agent is not required to eliminate or diminish massing of the massing polymer.

16. The process according to claim 14 wherein step (a) comprises,

- (1) feeding both the massing polymer and the ethylene- $\alpha$ -olefin at a ratio of 10-90 wt % ethylene- $\alpha$ -olefin to massing polymer into the feed throat of a twin screw extruder at a barrel temperature of 150-400°C;
- (2) optionally introducing free radical initiator and introducing an olefinic carboxylic acid or anhydride or derivative thereof into the extruder to form the polymeric toughening agent;
- (3) removing excess unreacted olefinic carboxylic acid or anhydride and isolating the polymeric toughening agent; and step (b) comprises,

15 melt-blending the polymeric toughening agent produced in step (a) with polyamide in an extruder, internal mixer or rubber mill at a temperature sufficient to melt the blend to form the polymeric composition.

17. The process of claim 16 wherein the polymeric toughening agent is selected from the composition of claim 1 and the olefinic carboxylic acid or anhydride is maleic anhydride and the polyamide is selected from nylon 6,6 or nylon 6.

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